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ELECTRICAL AND MAGNETIC PROPERTIES OF AMORPHOUS $Zr_{100-x}Cu_x$ ALLOYS

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The electrical resistivities and magnetic susceptibilities of seven amorphous $Zr_{100-x}Cu_x$ alloys ($26 < x < 71$) have been measured. A rather strong almost linear decrease of the Pauli like magnetic susceptibility with x is observed indicating that the electronic density of states at the Fermi level is dominated by Zr d-states in these alloys. Moreover these results can be simply explained by the dilution effect due to increasing Cu content. Their resistivities are high ($> 160\ \mu\Omega$ cm) and follow approximately the Nordheim rule with the maximum around $x = 60$. The resistivities of all alloys decrease continuously with temperature between 4.2 K and 300 K. The relative change in resistivity is bigger for the alloys with the lower Cu content. At low temperatures (but above the superconducting transition temperature, $T_c < 4$ K) their resistivities vary approximately as $-T^2$ and around room temperature a linear decrease of resistivity is observed. By the use of experimental structure factors the resistivities and the temperature coefficients of resistivity for three alloys (within our concentration range) have been calculated within the framework of Ziman's theory for liquid alloys. The comparison of the calculated and measured quantities shows that the above theory is not applicable to amorphous Zr-Cu alloys.

1. Introduction

The amorphous $Zr_{100x}Cu_x$ alloys can be regarded as model binary amorphous transition metal-metal system. The advantages of this system are that it is rela-

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tively simple and that the amorphous state can be obtained in a rather broad concentration (x) range. Despite this there is a limited number of data for this system and furthermore only a few physical properties have been measured in a broad concentration range.

Photoemission experiments[2,3] have shown that whereas the Zr d-band is rather close to the Fermi surface the Cu d-level has much higher binding energy. Thus the contribution of Cu is small and the electronic density of states ($EDoS$) near the Fermi level (E_F) is expected to resemble that of zirconium. Furthermore an increase in Cu content (x) is expected to decrease the $EDoS$ at E_F. A rather large electronic specific heat which decreases approximately linearly with x[4,5] supports that view. Preliminary measurements of the magnetic susceptibility on three ZrCu alloys[1,6] also support the above picture.

Recent measurements of the superconducting transition temperature (T_c) of these alloys[4,7] have shown that T_c's decrease approximately linearly with x as in $Zr_{100-x}Ni_x$ alloys[8]. This indicates that Zr d-electrons are responsible for the superconductivity of these alloys and also agrees[8] with the above picture of $EDoS$. Resistance measurements have shown that the resistivity decreases continuously with temperature and that the temperature coefficients of the resistivity (TCR) are negative over a broad concentration (x) range[7]. Such resistance variation was tentatively explained within the framework of Ziman's theory for the electrical resistivity of liquid alloys[9]. The problem was that resistance rather than resistivities were measured so that the actual x dependence of the resistivity was unknown.

In order to help the understanding of the physical properties of this system we have performed systematic investigation of the magnetic susceptibility and electrical resistivity of $Zr_{100-x}Cu_x$ alloys ($26 < x < 71$). Furthermore the electrical resistivity and TCR of three ZrCu aloys (spanning a broad x range) have been selfconsistently calculated[10] within the framework of the Ziman's theory. While the magnetic susceptibility can be successfully explained with the variation in $EDoS$ (described above) the calculated resistivity and TCR are at variance with the measured ones. The probable origin of this discrepancy is dicussed and the alternatives mentioned.

2. Experimental

The master alloys of predetermined concentration were prepared by arc melting of the high purity components in an argon arc furnace. The amorphous ZrCu alloys investigated in this study were prepared by melt-spinning[11] in air. The samples were ribbons 1—2 mm wide and 20—30 μm thick. All samples were investigated by x-ray diffraction before of other measurements. No trace of crystallinity was observed in any alloy. All measurements were performed on as obtained samples within few weeks after their production.

The magnetic susceptibility was measured on samples weighting few milligrams with the Faraday method[12]. The absolute values of the magnetic susceptibility were accurate to about one percent while the relative accuracy was about 0.1%. Since the magnetic susceptibility of two alloys ($Zr_{67}Cu_{33}$ and $Cr_{33}Bu_{67}$)

was practically independent of temperature ($80 < T < 300$ K) only the room temperature values were measured for the rest of the alloys.

The resistivity variation was measured on the samples of a few cm in length with the potentiometric technique[13]. The resolution of these measurements was 1 part in 10^5 over the entire temperature interval ($4.2 < T < 300$ K). Below 80 K the temperature was measured via calibrated Ge resistance thermometer and above with Pt resistance thermometer. The absolute resistivity values (determined via geometrical factor obtained from density, mass and length measurements) were accurate to about 2%. Deviations in resistivity equal or smaller than that amount were observed in measurements on several samples of the same alloy (but taken from different parts of the ribbon). The temperature coefficient of resistivity (*TCR*) was measured with the simple four probe technique between the ice point and room temperature (21 °C) achieved in suitable baths. The *TCR* values obtained on different samples of the same alloy agreed within a few percent. The uncertainty in *TCR* is not connected with the measurement technique but it is a consequence of the extreme sensitivity of this quantity on the actual preparation conditions[14].

3. Results and discussion

3.1. Magnetic susceptibility

The magnetic susceptibilities at room temperature of all our alloys are given in Table 1 both as measured and with ionic diamagnetic contribution subtracted (χ and χ_p, respectively). For this subtraction we used $\chi_{dia} = -12 \cdot 10^{-5}$ JT^{-2} mol^{-1} both for Zr and Cu[15]. The concentration dependence of the paramagnetic susceptibility (χ_p) is also shown in Fig. 1. Relatively large χ_p for the lowest x value (approximately equal to that of a pure crystalline Zr) and a rapid decrease of χ_p with x indicate that d-electron contributions to χ_p are dominant in these alloys[6]. Moreover on the basis of the observed x dependence of χ_p and of the results of the photoemission experiments (see introduction) it seems logical to assume that the only d-electron contribution to χ_p is due to Zr d-electrons. Thus, in a first approximation the effect of Cu on χ_p is dilution and χ_p of amorphous $Zr_{100-x}Cu_x$ alloys can be expressed as in paramagnetic transition metals[16]

$$\chi_p = \chi_o (1 - a\chi_o)^{-1} + \chi_{orb} \tag{1}$$

where $\chi_o = 2\mu_B{}^2 N_o$ (with μ_B the Bohr magneton value and N_o the bare density of states at E_F), the molecular field coefficient $a = I_{eff}/(2u_B{}^2 N_A)$ with I_{eff} the effective intraatomic exchange integral and N_A the Avogadro number and χ_{orb} is the orbital paramagnetic susceptibility.

From the specific heat results[4,5] the dressed electronic densities of states $N^* = (1 + \lambda)\ N_o$ (with λ the electron-phonon coupling parameter) and the Debye temperatures (Θ_D) for most of $Zr_{100-x}Cu_x$ alloys are known. From the experimental T_c values[4,7] by using the McMillan's expression[17]

$$T_c = \frac{\Theta_D}{1.45} \exp\left(-\frac{1.04\,(1+\lambda)}{\lambda - \mu^*(1 + 0.62\,\lambda)}\right) \tag{2}$$

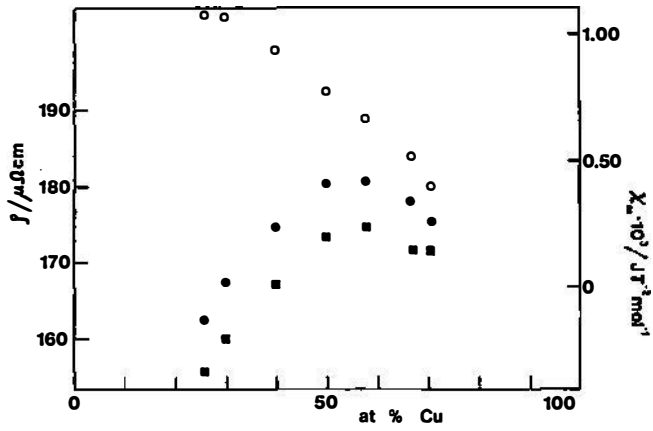

Fig. 1. Electrical resistivities at 4.2 K (●) and 273 K (■) and magnetic susceptibilities (○) of $Zr_{100-x}Cu_x$ alloys vs x.

with the effective Coulomb potential $\mu^* = 0.13$ we determined λ and hence N_t for all our alloys except $x = 71$ for which no specific heat results exist. The experimental T_c and Θ_D values as well as the calculated N_0 values are also given in Table 1.

TABLE 1

x in at%	$\chi \cdot 10^3$/ /(JT^{-2} mol^{-1})	$\chi_p \cdot 10^3$/ /(JT^{-2} mol^{-1})	T_c/K	N_0 (eV atom spin)	$a \cdot 10^{-2}$/J^{-1} T^2 mol
26	1.079	1.200	3.13	0.66	2.7
30	1.067	1.187	3.69	0.63	4.9
40	0.292	1.050	1.60[g]	0.57	5.8
50	0.775	0.895	0.70[g]	0.50	6.6
50	0.775	0.895	0.70[g]	0.50	6.6
58	0.658	0.778	0.3[g]	0.44	5.8
67	0.513	0.633	0.3[g]	0.40	6.7
71	0.384	0.514			

Θ_D/K	$\varrho_{4.2}$/μΩ cm	ϱ_{273}/μΩ cm	$\varrho^{-1}(\Delta\varrho/\Delta T)_{284} \cdot 10^4$/K^{-1}
192	162.3	155.4	—1.05
194[m]	167.1	160.0	—1.02
201[g]	174.4	167.2	—1.03
246[g]	180.1	173.2	—1.06
246[g]	180.1	173.2	—1.06
228[g]	180.4	174.4	—0.96
204[g]	176.2	171.5	—0.63
	175.2	171.5	—0.62

[m] Data taken from Ref. 5.
[g] Data taken from Ref. 4.

Data relevant to $Zr_{100-x}Cu_x$ alloys.

Since χ_p is practically independent of temperature one cannot obtain simultaneously a and χ_o from Eq. (1) for a single alloy[16]. Therefore we assumed that χ_{orb} of a given alloy is that of crystalline Zr ($97 \cdot 10^{-5}$ JT^2 mol^{-1}) scaled to the appropriate Zr content. This assumption is based on two facts:

i) χ_p of our alloys extrapolates to the value of a pure crystalline Zr when $x = 0$.

ii) photoemission experiments have shown that whereas Zr d-bands are close to Fermi surface, the Cu d-levels are at much higher binding energies.

Fitting our experimental χ_p values to Eq. (1) with χ_o and χ_{orb} determined as described above we obtained a values given in Table 1. It can be seen that a values increase a little with x and are within a scatter rougly equal to that of a pure crystalline Zr[16]. The exception is the alloy with $x = 26$ for which also the χ_p value was rather low compared to those of other alloys so that the corresponding a value should be regarded with caution. The above result can be regarded as a further evidence that $EDoS$ at E_F of amorphous Zr_{100-x} Cu_x alloys is dominated by Zr d--states.

3.2.. Electrical resistivity

In Fig. 1 the resistivities of all our alloys both at 4.2 K and 273 K are given. The resistivity in both cases follows the Nordheim rule with the maximum around $x = 60$. Within the investigated concentration interval the variation in resistivity with x is rather weak (ten percent or less of the total resistivity). The resistivity values are consistently high for all alloys (> 155 $\mu\Omega$ cm) while their variation with temperature is rather small as seen from the difference between the data points for 4.2 and 273 K. The resistivities both at 4.2 and 273 K are also given in Table 1.

The resistivities of amorphous alloys are usually described in terms of Faber--Ziman theory[9]. Since the original Ziman's theory was not applicable to the strong scattering case the theory was extended to describe also liquid transition metals and their alloys. In this calculation[18] the actual d-band of the transition metal is replaced with a rather narrow resonance of a width Γ. The resistivity than depends strongly on the relative position of E_F in respect to the peak of the resonance. Apparently with suitably selected E_F (hence k_F) the theory was able to obtain semi-quantitative agreement with a number of experimental results.

More recently the most complete discussion of extended Ziman's theory[18] has been given[10]. It was pointed[10] that $2k_F$ should be calculated in the same spirit as the resistivity itself i. e. by using Loyd's formula for the integrated density of states[19]:

$$N(E_F) \cong N_o(E_F) + \frac{2}{\pi} \sum_l (2l+1) [c_1 \eta_l^{(1)}(E_F) + c_2 \eta_l^{(2)}(E_F)] \qquad (3)$$

where $N_o(E_F)$ is the integrated density of states for free electrons, η_l are the phase shifts and c_1 and c_2 are the concentrations of the constituent metals. E_F (hence k_F) is to be determined from the condition $N(E_F) = c_1 Z_1 + c_2 Z_2$ with Z the total number of valence electrons (in our case $Z = 4$ and 11 for Zr and Cu, respectively).

Since we obtained the reliable resistivity data of Zr_{100-x} Cu_x alloys in a broad concentration range it was of interest to compare the experimental values with those calculated on the basis of Faber-Ziman theory. In this theory the resistivity of a binary alloy is given with the following expression:

$$\varrho = \frac{12\,\pi\,\Omega_o}{e^2\,h\,v_F}\int_0^{2k_F} dq\,q^3\,\langle T_{alloy}\rangle^2 \tag{4}$$

where

$$\langle T_{alloy}\rangle^2 = c_1\,(t_1)^2\,(1 - c_1 + c_1\,a_{11}\,(q)) + c_2\,(t_2)^2\,(1 - c_2 + c_2\,a_{22}\,(q)) +$$

$$+ c_1\,c_2\,(t_1^*\,t_2 + t_1\,t_2^*)\,(a_{12}\,(q) - 1)$$

and

$$t = -\frac{2\,\pi\,h^3}{m\sqrt{2m\,E}\,\Omega_0}\sum_l (2l + 1)\sin\eta_l\,(E)\,e^{i\eta_l(E)}\,P_l\,(\cos\Theta)$$

here $\Omega_0 = c_1\,\Omega_{01} + c_2\,\Omega_{02}$ is the atomic volume, v_F is the Fermi velocity, Θ is the scattering angle and a_{ij} are the structure factors.

In calculating the resistivity we determined $2k_F$ from Lloyd's formula (Eq. 3) and used the experimental structure factors[21] and published phase shifts[20]. The calculation was simplified by taking only d-resonant terms in the resistivity expression (Eq. 4). The results of this calculation for three Zr_{100-x} Cu_x alloys (for which the experimental structure factors were known) are given in Table 2 together with some important parameters used in this calculation. Comparing these values with the experimental ones (Table 2, 4 th column and also Table 1) it can be seen that the calculation neither gives the magnitude nor the concentration (x) dependence of the resistivity correctly. As regards the magnitude of the resistivity we note that (like extended Ziman's formula) the multiple scattering term was neglected in this calculation. Some attempts to include the short range order effect in this theory (within the quasicrystalline approximation) resulted[22] in a decrease in the calculated resistivity values. However the problem of applicability of the Boltzmann equation in such systems remains unsolved in this approach. For this reason we would not expect good agreement between the calculated and observed resistivites even if the calculation were improved in the above[22] manner.

TABLE 2

x in at%	E /eV	$2k_F \cdot 10^{-10}$ m	$\varrho_{calc}/\mu\Omega$ cm	$\frac{\varrho_{calc} - \varrho_{exp}}{\varrho_{exp}}$	$\varrho^{-1}\left(\frac{\Delta\varrho}{\Delta T}\right)_{R.T.} \cdot 10^4/K^{-1}$
65	7.875	2.88	280	0.57	0.75
50	7.738	2.85	350	0.94	1.50
35	7.657	2.82	420	1.48	2.07

Calculated resistivities and temperature coefficients of resistivity at room temperature for three Zr_{100-x} Cu_x alloys.

The temperature dependence of the resistivity of $Zr_{100-x}Cu_x$ alloys is shown in Fig. 2. Here the normalized resistivity (normalized to its value at 4.2 K) is plotted in order to emphasize the eventual similarity in the resistivity variation for the alloys with different x. Indeed the resistivity variation looks similar in all the alloys and there is a definite decrease in the change of resistivity from 4.2 to 300 K with increasing x. The relative change in resistivity from 4.2 to 273 K, $(\varrho_{4.2} - \varrho_{273})/\varrho_{4.2}$, for all our alloys is shown in Fig. 3. The resistivity variation around room temperature looks linear in all alloys. This allows the determination of the temperature coefficient of resistivity $\alpha = \varrho^{-1}(\Delta\varrho/\Delta T)$ at 284 K which is also shown in Fig. 3. It can be seen that except for different sign the variations of α and $(\varrho_{4.2} - \varrho_{273})/\varrho_{4.2}$ with x are practically the same. α is negative throughout the explored concentration range and tends to smaller values at Cu rich end. The negative α values are consistent with the empirical correlation established by Mooij[23] according to which the alloys with the resistivity higher than 150 $\mu\Omega$ cm have negative α values.

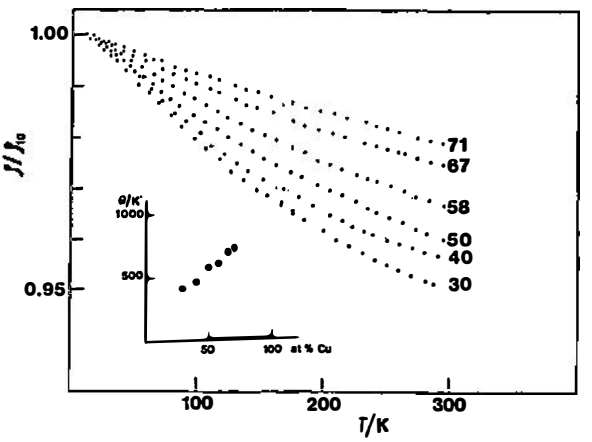

Fig. 2. Normalized electrical resistivities of $Zr_{100-x}Cu_x$ alloys vs temperature. Numbers denote x. In the inset: the characteristic temperatures Θ obtained from the low temperature resistivity (see text) vs x.

In Ziman's theory both the resistivity and the temperature coefficient of resistivity (α) are strongly related to the structure factor. Following Nagel[24] we estimated α for the same three Zr Cu alloys for which the resistivities were calculated. These calculated α values are also given in Table 2. It can be seen that both the sign and x dependence of the calculated α values are at variance with the observed ones (Fig. 3).

At low temperatures (but above the superconducting transition temperature) the resistivity of all our alloys tends to saturation. We found that at these temperatures ($4.2\ \mathrm{K} < T < 50\ \mathrm{K}$) the resistivity can be fitted with a reasonable accuracy to the empirical relation:

$$\varrho = \varrho_0 \left(1 - \left(\frac{T}{\Theta}\right)^2\right) \tag{5}$$

where ϱ_0 is the resistivity value at its maximum ($T \sim 4.2$ K) and Θ is a constant for a given alloy obtained from the fit of experimental data to Eq. (5). In the inset to Fig. 2 the values of Θ are plotted as a function of x. It can be seen that Θ values increase (approximately linearly) with x.

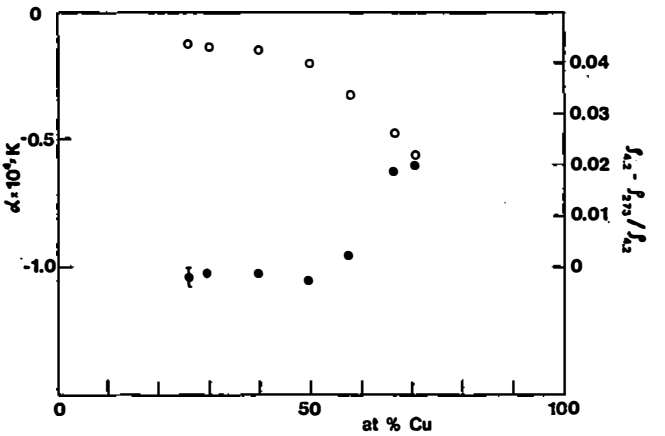

Fig. 3. Relative change in resistivity (●) and the temperature coefficient of resistivity, $\alpha = \varrho^{-1}(\Delta\varrho/\Delta T)_{284K}$ of $Zr_{100-x}Cu_x$ alloys vs x.

The low temperature resistivity can also be calculated within the framework of the Ziman's theory. By taking into account the inelastic scattering effects Cote and Meisel[25] get unavoidably $a + T^2$ increase of the resistivity at low temperatures. Apparently this result also contradicts the observed behaviour (Fig. 2). Moreover the comparison of our Θ values with the experimental Debye temperatures (Table 1) seems to rule out the phonon effects as the origin of the low temperature resistivity variation.

The above discussion indicates that the Ziman's theory (when applied self-consistently) predicts wrong concentration and temperature dependence for the resistivity of amorphous $Zr_{100-x}Cu_x$ alloys. A wrong concentration dependence may possibly be caused by the neglect of d-electron contribution to the conductivity. Although it is generally true that in transition metals and alloys d-electrons are responsible for their superconducting and magnetic properties while their contribution to conductivity is negligible, the situation may be somewhat different in amorphous alloys. In the case of strong structural disorder (short electronic mean free path) the contribution of d-electrons to conductivity may not be negligible. This seems to be the case in amorphous Zr-alloys where the electronic mean free path is of the order of a few interatomic distances and where the effective mass of d-electrons may not be so large due to rather broad d-band. Furthermore the Hall coefficients of amorphous Zr alloys are invariably positive[26] which rules out the applicability of free electron picture to these alloys.

A wrong temperature dependence of the resistivity predicted by the above model indicates that the more recent concepts for the resistivity of disordered solids such as incipient localization, the influence of the electronic interaction on

the scattering of electron on disorder and the electron scattering on unstable ionic configurations (defects) may be useful in understanding the resistivity of these and similar amorphous alloys.

4. Conclusion

Accurate data for the room temperature magnetic susceptibility and the concentration and temperature dependence of the resistivity of amorphous $Zr_{100-x}Cu_x$ alloys ($26 < x < 71$) have been presented. The magnetic susceptibilities of these alloys are compatible with their *EDoS* deduced from photoemission experiments. Moreover the experimental values can be reproduced from Eq. (1) by the use of experimental N_0 values (obtained from specific heat measurements) and under the assumption that the orbital contribution to the magnetic susceptibility is that of a pure Zr, scaled to the appropriate Zr content. This procedure yields the effective intraband Coulomb interaction practically the same as in a pure Zr lending further support to the experimentally observed *EDoS*.

The variation of resistivity with x is qualitatively the same both at 4.2 and 273 K. In both cases the resistivity exhibits a shallow maximum around $x = 60$. This x dependence of resistivity cannot be explained within the Faber-Ziman theory if the calculation is performed selfconsistently and the experimental structure factors are used. The temperature dependence of resistivity is qualitatively the same for all alloys: there is a $-T^2$ variation below 50 K and approximately $-T$ dependence around room temperature. The variation of resistivity with temperature becomes less pronounced with increasing x. The above temperature dependences of the resistivity are also at variance with those predicted within the framework of Ziman's theory. It is conjectured that the neglect of d-electron contribution to conductivity may be one of the reasons for the inapplicability of Ziman's theory to amorphous ZrCu alloys.

During preparation of the manuscript an important paper escaped our notice. Gallagher and Greig[26] measured and calculated thermoelectric power and resistivity of several amorphous ZrCu alloys. Their experimental room temperature resistivity values agree well with our ones. They also calculated the resistivities for three alloy in terms of extended Ziman model and obtained a result very similar to our one. Furthermore they also calculated the resistivities in terms of Mott's model[27] and obtained a similar disagreement with the experimental results. This is not surprising since both Ziman's and Mott's model are similar in the way that only s-conduction is taken into account.

Recently we have been informed by Dr. P. Rhodes[28] that he performed a similar analysis of the magnetic susceptibility results of some glassy ZrCu alloys. He also concluded that the orbital contribution to the magnetic susceptibility is dominant in these alloys. However due to somewhat smaller orbital magnetic susceptibility of Zr used in this work he obtained considerably higher enhancement factor I_{eff}.

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ELEKTRIČNA I MAGNETSKA SVOJSTVA AMORFNIH $Zr_{100-x}Cu_x$ SLITINA

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Mjereni su električni otpor i magnetska susceptibilnost sedam $Zr_{100-x}Cu_x$ slitina ($26 < x < 71$). Jako, gotovo linearno smanjenje Paulijeve magnetske susceptibilnosti s x ukazuje da je gustoća stanja elektrona na Fermijevom nivou u tim slitinama dominirana Zr d-stanjima. Štoviše rezultati se mogu jednostavno objasniti

efektom razrijeđenja povećanjem koncentracije Cu. Električne otpornosti su visoke ($> 160\ \mu\Omega$ cm) i približno slijede Nordheimovo pravilo s maksimumom oko $x = 60$. Otpornosti svih slitina kontinuirano opadaju s temperaturom između 4,2 i 300 K. Relativna promjena otpora je veća za slitine s manje Cu. Na niskim temperaturama (ali iznad temperature supravodljivog prijelaza, $T_c < 4$ K) otpori slijede $-T^2$ ovisnost dok oko sobne temperature opadaju linearno s temperaturom. Korištenjem eksperimentalnih strukturnih faktora električne otpornosti i temperaturni koeficijenti otpornosti na sobnoj temperaturi su izračunati za tri slitine (unutar našeg područja koncentracije) pomoću Zimanovog modela. Usporedba izračunatih i mjerenih vrijednosti ukazuje da taj model nije primjenljiv na amorfne ZrCu slitine.